TESTING AND EVALUATION OF AN ADVANCED HIGH PERFORMANCE PLANAR SOFC STACK

FINAL PROGRESS REPORT

- **PREPARED BY:**

  SOFCo
  2425 South 900 West
  Salt Lake City, UT 84119

- **PREPARED FOR:**

  U.S. Army Research Office
  P. O. Box 1211
  Research Triangle Park, NC 27709-2211

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TABLE OF CONTENTS

FIGURES

1.0 INTRODUCTION

Planar SOFC technology development at SOFCo, a McDermott Technology Inc./Ceramatec partnership, has the primary objective of demonstrating compact, high efficiency power generation systems with multi-fuel capability meeting commercial performance goals. SOFCo has conducted several programs which synergistically address this objective: an internally funded program focusing on stack development and system integration for pipeline natural gas (PNG) operation, a DARPA sponsored program to demonstrate logistic fuel operation, and a program to develop cell and stack technology for low temperature operation supported by EPRI.

With sponsorship from DARPA and the U.S. Army, SOFCo concluded work on a three-phase, four-year program to demonstrate a mobile electric power (MEP) fuel cell generator operating on logistic fuel. The primary aim of the program was to integrate planar solid oxide fuel cells with a JP-8 fuel processor into a compact generator module. The integration of partial oxidation fuel processor for reforming high sulfur (0.3% by weight) logistic fuel, with SOFC stacks into a compact hardware configuration has the potential to advance the state-of-the-art in SOFC system technology.

Significant advances were made in all aspects of the SOFC development activities conducted in Phase 3 of the MEP program and supporting internal programs. These advances include increased cell power density, stack power density, stack lifetime, cell manufacturing capability, stack height, and seal effectiveness. This enabled demonstration of a 1-kW JP-8 fueled SOFC system in the technology readiness review (TRR) test which achieved the 1997 program performance objectives for initial power density (ASR ~ 2.5 -cm^2) and fuel utilization (50%). While the degradation rate observed in the TRR test was reduced several fold relative to the previous year, it was still too high to meet lifetime objectives (Figure 1).
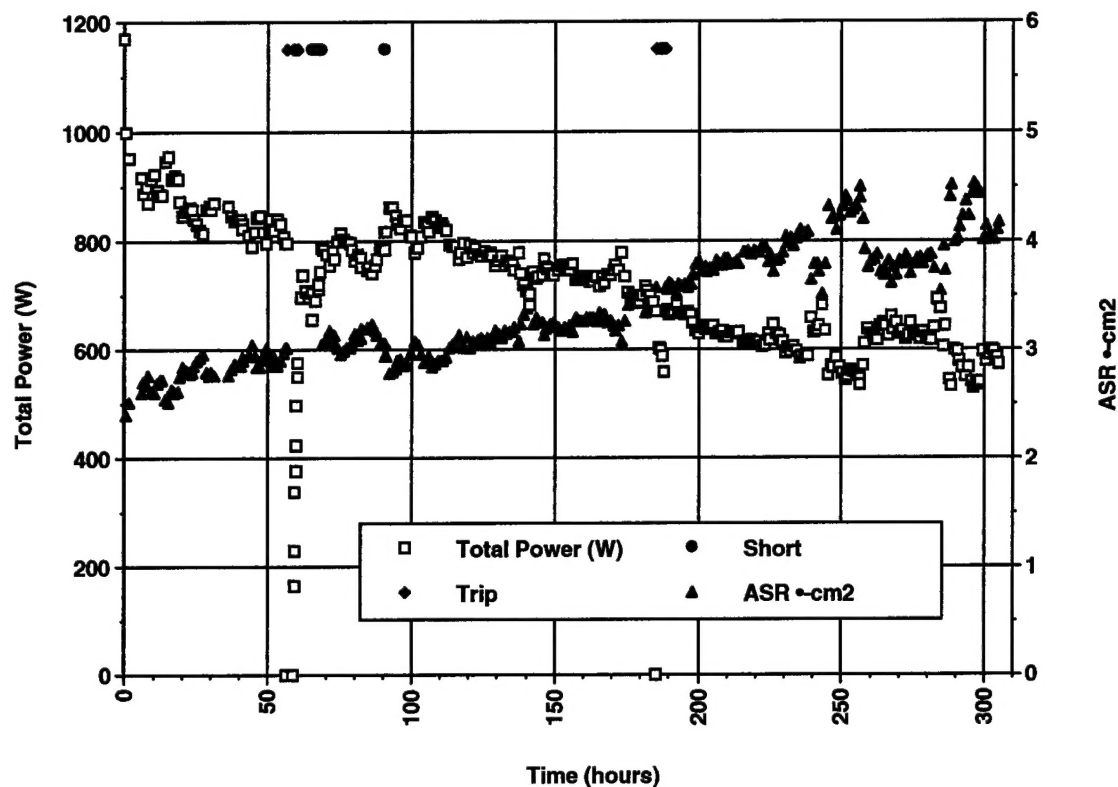
Figure 1. TRR Stack Performance with Time.

The two stacks in the multi-stack TRR test unit exhibited similar performance characteristics and closely matched performance predictions. The major issue was high stack degradation. This high degradation was not unexpected and was a consequence of using commercial stainless alloy interconnects. These were used due to limited availability of a specially designed proprietary K30 alloy and limited experience with an advanced metal interconnect design. A small stack using this new light weight interconnect design has demonstrated stable high performance operation over 3500 hours, as shown in Figure 2. Intensive development on an aggressive, internally funded program to scale this technology to TRR size stacks is underway.

Additionally, SOFCo utilizes a unique Ni-cermet anode catalyst which offers high Sulfur tolerance. This was demonstrated in the MEP program by successful testing of single cells for more than 4,000 hours with 1,000 ppm of sulfur in fuel.

2.0 <u>METAL INTERCONNECTS</u>

In contrast to conventional monolithic interconnects, SOFCo has developed a new advanced interconnect. This advanced design has been employed successfully at the 5-cm size, and has resulted in the stack performance shown in Figure 2. This design employs a novel materials set and an innovative design which offers a significant weight reduction to the stack.
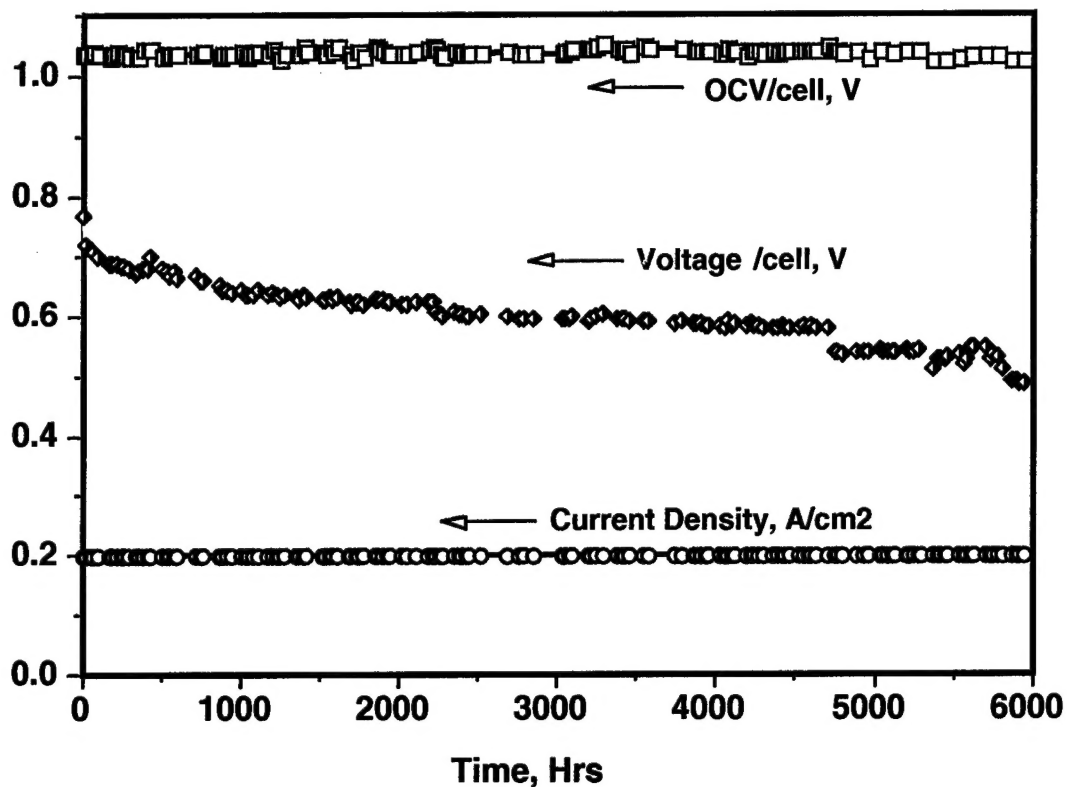
Figure 2. Advanced Interconnect Stack Performance.

3.0 <u>SULFUR TOLERANCE</u>

The issue of sulfur tolerance was addressed at the single cell level. Cell performance at various concentrations of sulfur in fuel was tested. Negligible effect was seen with 5 ppm H_2S and a small drop in performance was observed with 50 and 500 ppm sulfur in fuel (Figure 3). Cells were tested for up to 2,000 hours with hydrogen fuel containing 50 ppm and 500 ppm of H_2S. The effect of 50 ppm H_2S was negligible, with no additional degradation observed. With H_2S concentrations of 500 ppm, a reversible 50mV drop was observed, though no degradation with time was observed in 2000 hours of operation. Sulfur concentrations as high as 1,000 ppm did not show much instability in performance. The long term test results using different H_2S concentrations in the fuel are shown in Figures 4 - 6. This is encouraging to find that the cell component is relatively unaffected by sulfur though it is unlikely that metal interconnects and process piping will fare as well as the ceramic cell. The 50 ppm tested is in the range of measured H_2S concentrations from the POx system. Temporary exposure at this concentration due to a saturated ZnO bed or malfunctioning bypass would be expected to have little if any effect on the stacks.
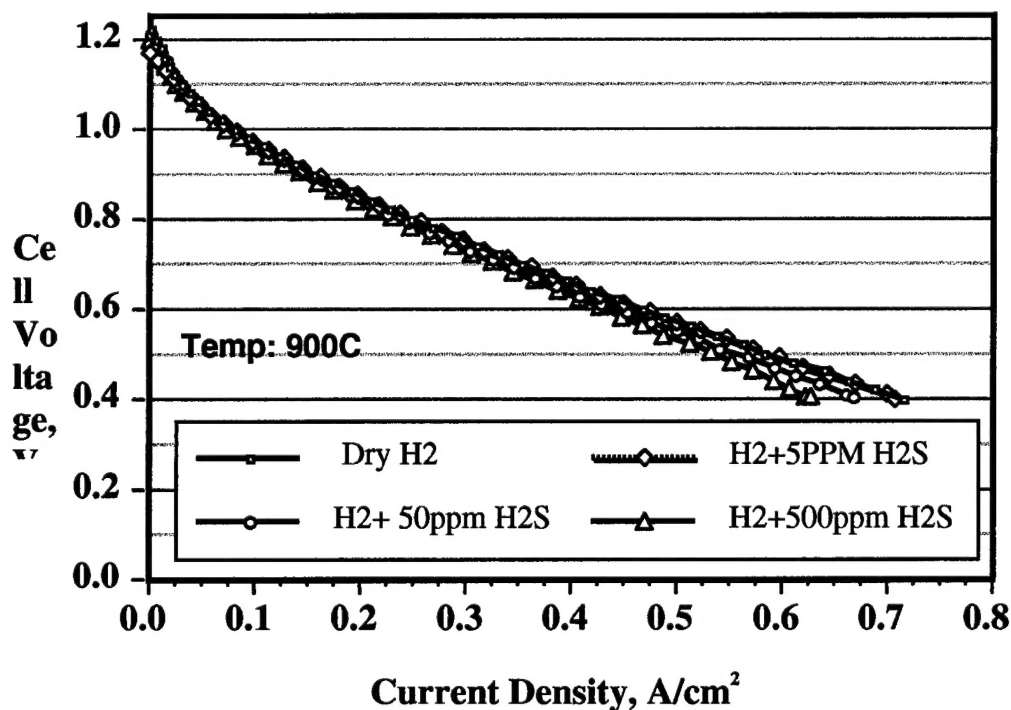

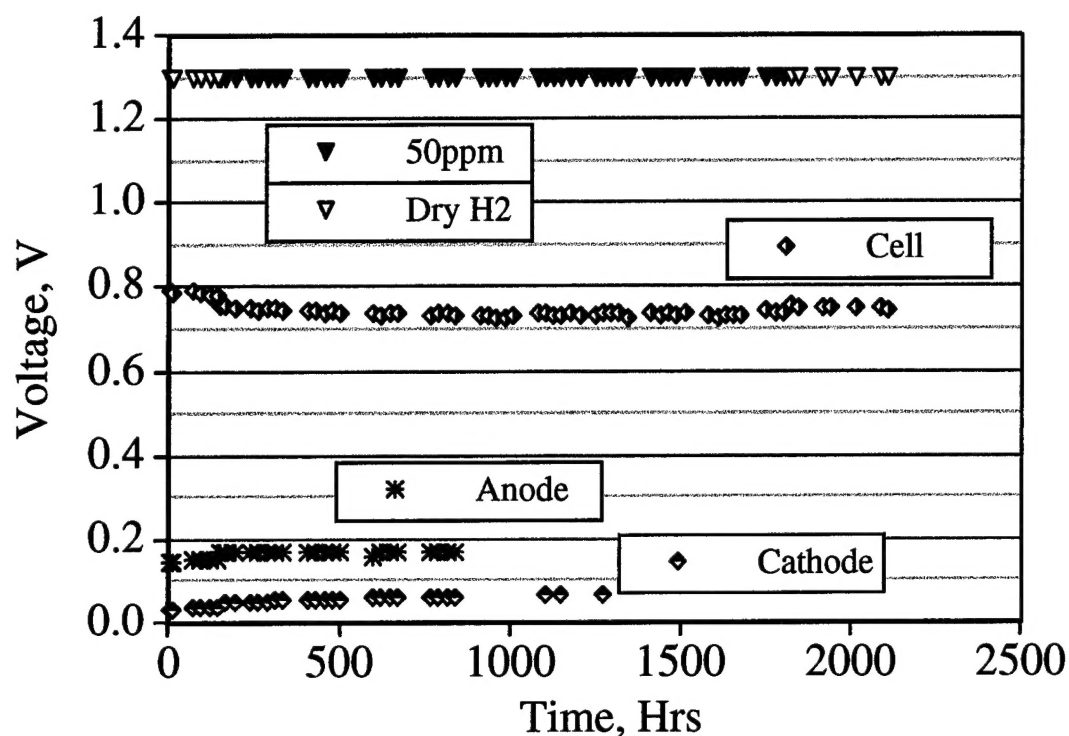**Figure 3. Effects of Sulfur in Fuel on Cell Performance.**

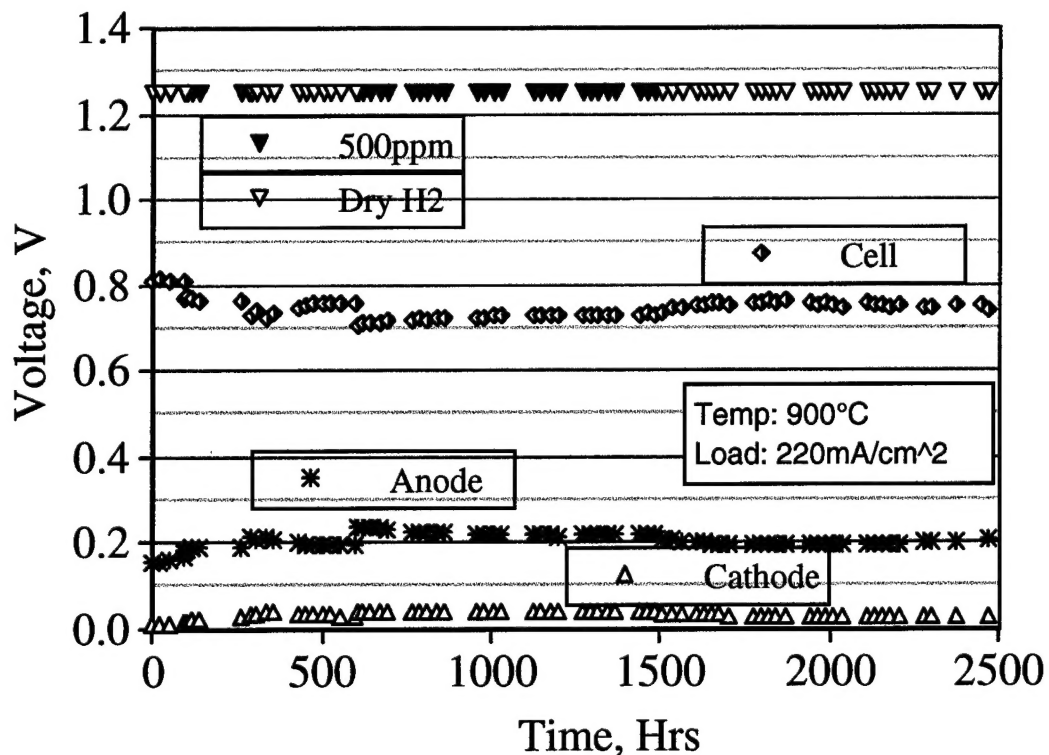**Figure 4. Long Term Performance with 50 ppm Sulfur in Fuel.**

Figure 5. Long Term Performance with 500 ppm Sulfur in Fuel.
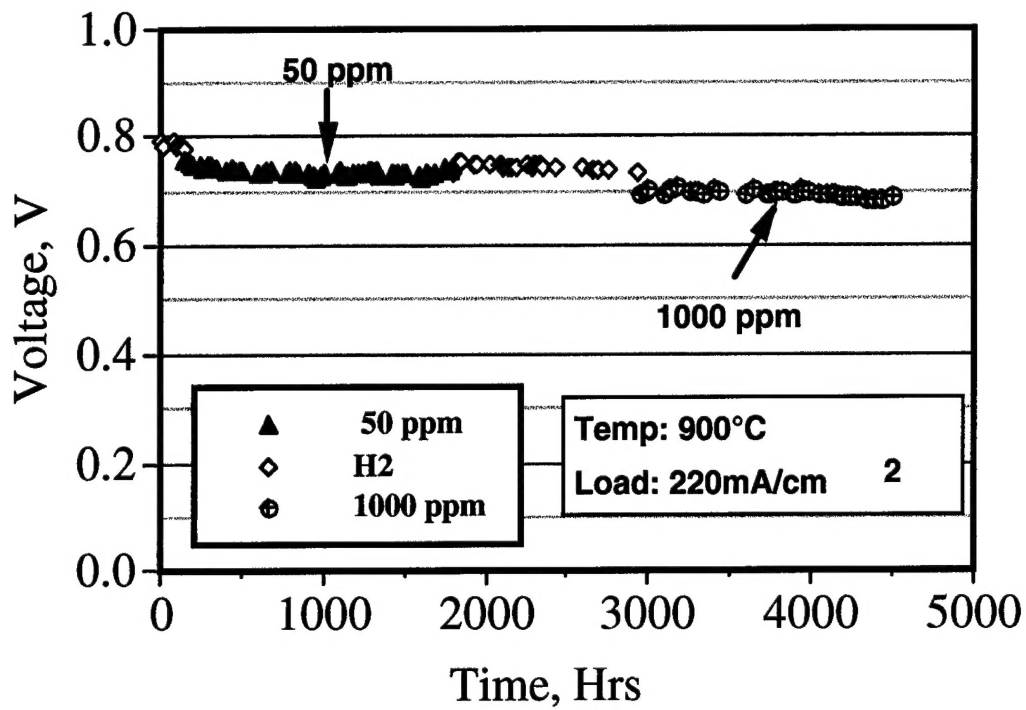
Figure 6. Long Term Performance with 1,000 ppm Sulfur in Fuel.

4.0 <u>TECHNICAL PROGRESS</u>

Based on the results from the short stack endurance test with the advanced interconnects and the sulfur tolerance tests using single cells, the current program focused on the following tasks: testing of short stacks using advanced cell concepts, sulfur tolerance testing of short stacks, and establishing stable tall stack performance. Since April '98, the primary focus at SOFCo has been to scale up the advanced metal interconnect stack results to 10 cm size and to lower degradation rates using improved surface treatments for the metal interconnects.

4.1   *Task 1: Short Stack Fabrication and Testing:*

The primary objective of this task is to test 10x10 cm short stacks using advanced metal interconnects and establish process parameters to demonstrate stability similar to the 5x5 cm short stacks. Several short stacks (4 cells) were tested to investigate the following parameters: cell size effect (performance of 10 cm stacks relative to 5 cm stacks), stability of short stacks as a function of interconnect material and surface treatment process. The initial ASR of the stacks ranged from 1.7 to 2.4 ohm.cm^2 at an operating temperature of 850°C. Most stacks showed an initial increase in resistance during the first 50 hours of operation. Two stacks showed an increase of only 0.3 ohm.cm^2 before exhibiting stable performance. These two stacks performed with a nearly constant degradation rate for a test duration of ~2500 hours. Stack #364 has similar stability to the target performance of stack #337. This is a marked difference compared to the MEP-TRR stack. Figure 7 shows the performance of these stacks.

Several single cell tests were performed to qualify the electrode inks. Cells showed that the electrodes are capable of providing a cell performance of ~1.5 ohm.cm^2 at 850°C. Multiple short stacks using 10 cm components were also tested to qualify stacking process. The stacks were consistent in reproducing the starting ASR of 2.5 ohm.cm^2 at an air inlet temperature of 770°C. Some of the stacks however showed lower OCV than expected. Since the short stacks exhibited lower OCV a 20-cell stack was built using actual components to determine whether the stacking process introduced cracks in the electrolytes. Disassembly of the stack showed only one electrolyte

These short stack tests served as qualification tests for process variables leading to successful tall stack tests. During the program period several short stacks and three tall stack tests were conducted. Tall stack units 2 and 3B were tested under the internal program, and stack 3A was tested under the DARPA program.
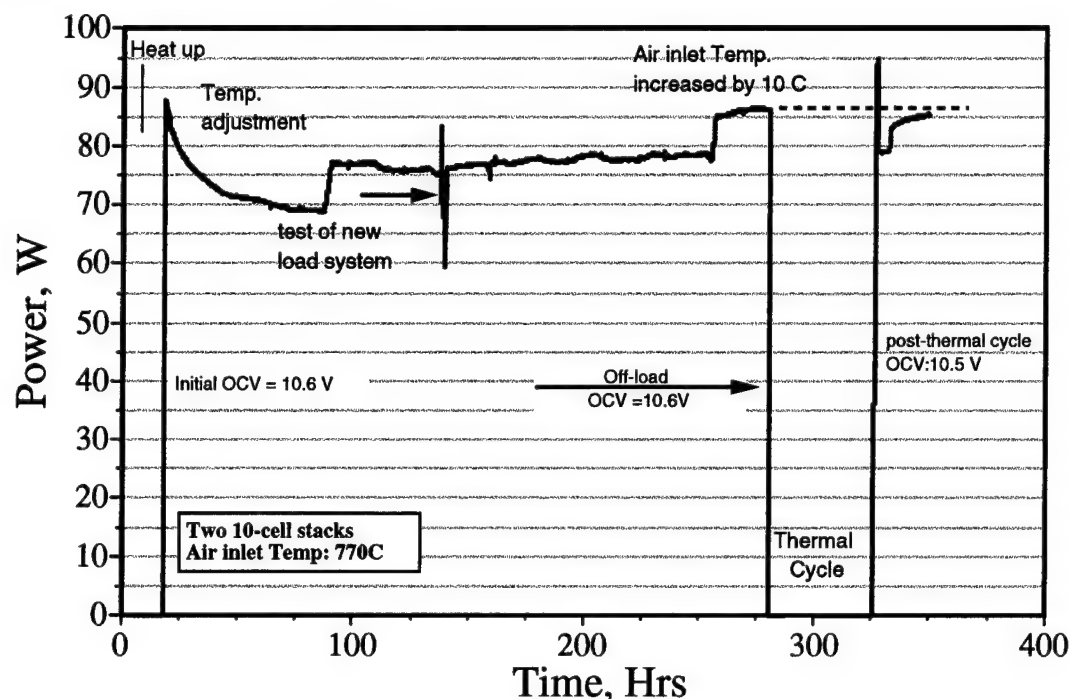
Figure 8. Performance of Short Stacks.

4.2   *Task 2: Fabrication and testing of a 60 cell stack:*

The objective of this task is to scale the advances made in the short stacks with advanced interconnects to tall stacks. A tall stack is defined as having approximately 50 cells of 10x10 cm size. The commercial target is a 0.25% drop in voltage per 1000 hours, after an initial burn-in period. If the loss in voltage were linear with time, this voltage drop would lead to a stack lifetime of 40,000 hours with an acceptable voltage drop of 10%. This task was aimed at the fabrication, testing and evaluation of a tall 10 cm stack for a period of ~ 500 hours. The results will be compared with results from analytical predictions and historical data such as the TRR test results. This task was supported by the ongoing activity in SOFCo on the internal program.

A tall stack (Unit 1) consisting of 46 cells (10x10cm) was tested in July '98 using SOFCo iternal funding. The starting performance showed an ASR of 2.3 ohm.cm^2 as expected from short stack results. The ASR increased with time, following a similar trend exhibited by short stacks of identical cells and process techniques.

The second tall stack (Unit 2) consisting of two 10x10cm 40 cells stacks was tested in August '98 using SOFCo internal funding. This unit has operated for over 2000 hrs and has shown the best tall stack performance and stability to date. The interconnects used in this stack have an improved oxidation resistant coating, which has been shown in ex-situ tests to exhibit a 50% lower resistance and improved stability. Compared to the December '97 "MEP-TRR " test conducted using monolithic stainless steel interconnects, this stack pair showed an order of magnitude improvement in degradation rate. The degradation rate of the MEP-TRR test was ~ 300% / 1000 hr while the degradation rate of Unit # 2 is ~ 28%/1000 hrs. In comparison to the previously demonstrated short stack test milestone of ~ 5%/1000 hours, considerable improvement needs to be made. It is anticipated that this will occur when system related upset conditions, like those seen during the stack operation, which cause precipitous loss in stack power, are avoided. By making changes to the test station, chances of such upsets repeating are expected to be reduced.

A third tall stack consisting of 80 cells of 10-cm components was assembled. The unit (designated 3A) was heated to 750°C air inlet temperature. The open circuit voltage was lower than theoretical and the diagnostic leads indicated that some segments of the stacks are shorted. A diagnostic test of the unshorted segments showed an ASR of ~2.5 ohm.cm^2, similar to the previous tall stack and the short stacks. The stack was cooled down and inspected. Several of the cells were found to have shorted to the manifold due to inadequate application of the dielectric. The manifolds were removed and replaced by another set and the stack retested. Some of the segments still had lower lower OCV and thus the stack was cooled again and the lower OCV segments were bypassed for the next heat up. The segments that were operational showed near theoretical OCV despite two complete thermal cycles and a manifold replacement. The stack was operated for more than 350 hours. The performance of the stack was stable after an initial improvement in performance as shown in Figure 9.
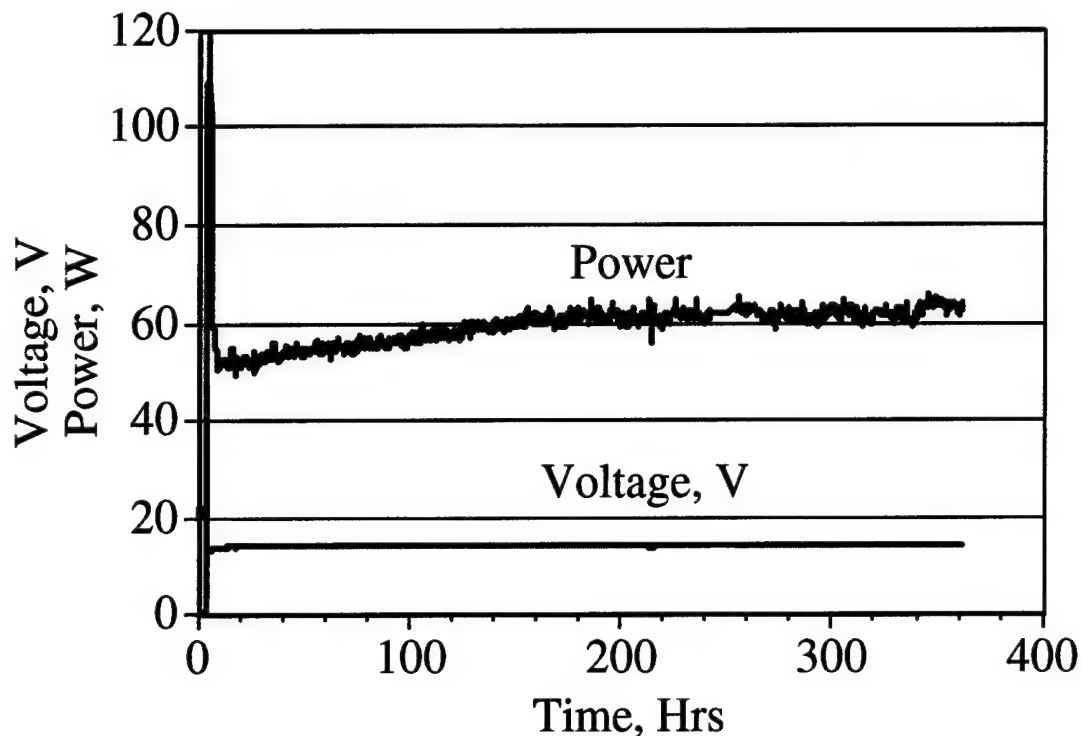
Figure 9. Performance of Unit 3A.

4.3   *Task 3: Evaluation of S Tolerance of Short Stacks:*

The primary objective of this task is to test short stacks with S containing fuel. In order to obtain unambiguous stack performance results without the fuel feed piping consuming the S in the fuel through chemical reactions, fuel feed modifications of a short stack test station was first completed. To shakedown the feed system modification, a 10 cell 10x10cm stack with commercial stainless steel interconnects (TRR vintage) was installed. The stack was tested using the normal procedures, and after determination of its performance characteristics and stability, 50ppm H_2S was introduced into the fuel feed. After an initial drop in performance, the stack showed a steady degradation rate consistent with expectations from the interconnects of the selected composition. Switching the fuel back to H_2 increased the stack current, as expected. As the fuel was switched to

50ppm H_2S the current - time trace followed the extrapolated curve for the same fuel composition (Figure 10). The high degradation of these interconnects amplifies the effect of S in the fuel.
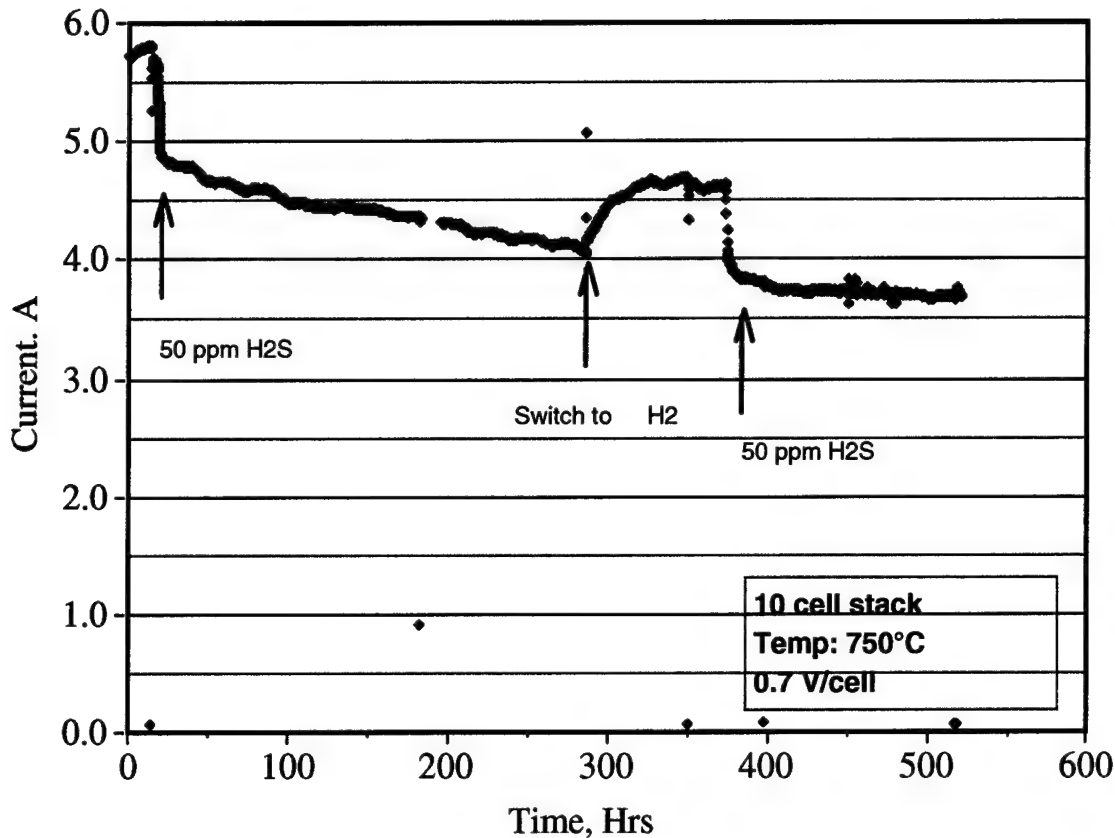
**Figure 10. 500-hr Performance of a Short Stack in S containing Fuel
Using commercial Stainless Steel Interconnects**

The short stack (10x10 cm, 10 cells) using stainless steel interconnect was terminated after 1200 hours. The endurance characteristics of this stack is illustrated in the power degradation plot shown in Figure 11 . The degradation rate of this stack was 36 %/1000 hr with 50 ppm S in the fuel. In comparison, the degradation rate as seen from the graph appears to be 29 %/1000 hrs without S in the fuel. This increase in degradation rate is likely an effect of scaling when, on introducing S in the fuel feed, a portion of the active electrode is covered with S resulting a decrease in power output. The absolute magnitude of the change in power output for both S loaded and S free fuel conditions is similar.
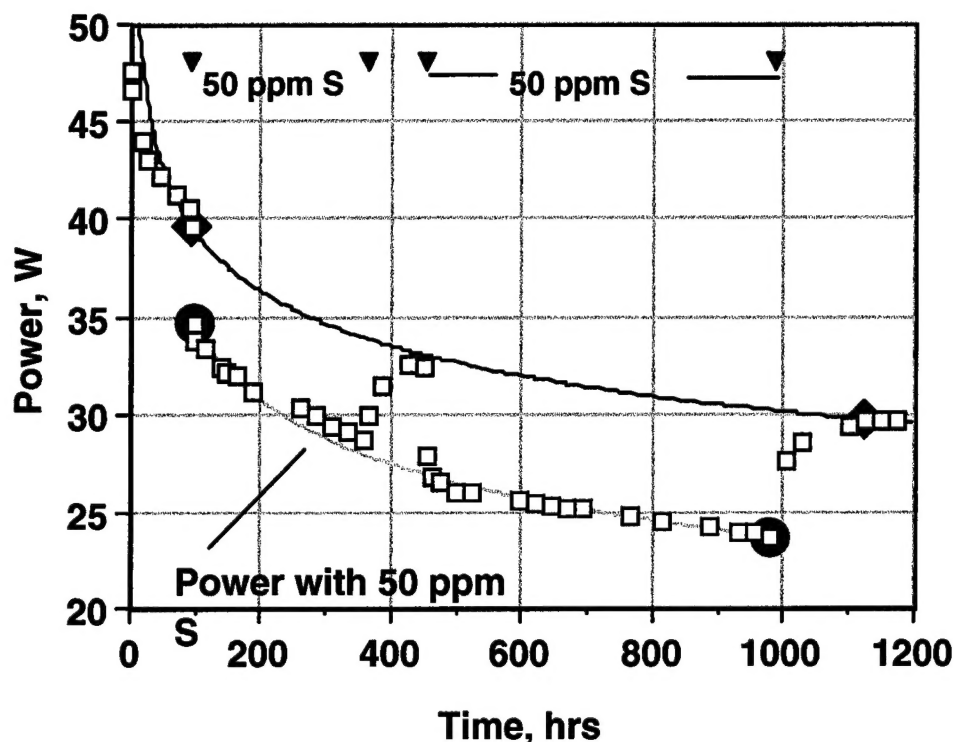
Figure 11: Endurance Characteristics of a 10 cell, 10 cm Stack, Using Commercial Stainless Steel Interconnects.

A second short stack was tested using the advanced metal interconnects. Since the stack had cells of different variations only cells comparable to the previous test are reported. In order to directly compare the two stacks, normalized ASR units are used in Figure 12. The figure shows both the stainless steel interconnect stack (top trace) and the advanced metal interconnect stack (bottom trace). It can be seen that the advanced stack shows both lower ASR and lower degradation. The ASR is slightly higher with S containing fuel than the hydrogen fuel. The degradation rate is nearly the same with S containing fuel as the hydrogen fuel until the 600 hr period after which the degradation appears to increase. The cause of this increase is not clear at present. Additional tests are required to establish a clear trend. The results are however encouraging that in general the stack performance is only slightly affected if a S slip occurs and that even to extended exposure the degradation trend is similar to S-free fuel operation.
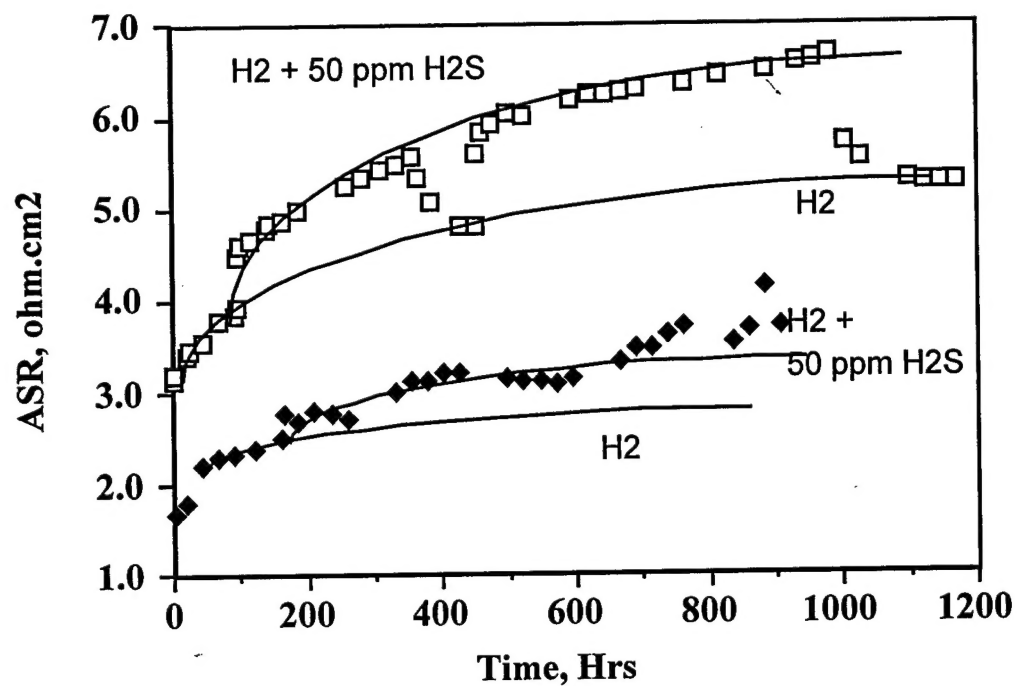
Figure 12. Comparison of Short Stack Performance in S containing Fuel.

(Commercial stainless steel interconnects - top trace)

5.0 <u>CONCLUSIONS</u>

The primary objectives of the program were to establish stable operation in tall stacks and demonstrate sulfur tolerant operation of short stacks. Tall stacks tested under both internally funded and DARPA funded programs demonstrated significant progress toward achieving these goals. The tall stacks tested during the program period demonstrated a stability which was an order of magnitude better than the MEP-TRR test. Many of the instabilities in performance are traced to system related instabilities and steps have been taken to address those issues since the conclusion of this program. Short stacks using the advanced metal interconnects demonstrate a small drop in cell voltage with the presence of sulfur (in H_2S form) and no appreciable change in stability over time. Thus a sulfur slip in the fuel is not expected to drastically affect the stack performance or endurance.

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Dr. S. Elangovan

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SOFCo   McDermott Technology, Inc.
2425 South 900 West   1562 Beeson Street
Salt Lake City, UT 84119   Alliance, Ohio 44601

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13. ABSTRACT (Maximum 200 words) Planar SOFC technology development at SOFCo, a McDermott Technology Inc./Ceramatec partnership, has the primary objective of demonstrating compact, high efficiency power generation systems with multi-fuel capability meeting commercial performance goals. SOFCo has conducted several programs which synergistically address this objective: an internally funded program focusing on stack development and system integration for pipeline natural gas (PNG) operation, a DARPA sponsored program to demonstrate logistic fuel operation, and a program to develop cell and stack technology for low temperature operation supported by EPRI. The primary objectives of the program were to establish stable operation in tall stacks and demonstrate sulfur tolerant operation of short stacks. Tall stacks tested under both internally funded and DARPA funded programs demonstrated significant progress toward achieving these goals. The tall stacks tested during the program period demonstrated a stability which was an order of magnitude better than the MEP-TRR test. Many of the instabilities in performance are traced to system related instabilities and steps have been taken to address those issues since the conclusion of this program. Short stacks using the advanced metal interconnects demonstrate a small drop in cell voltage with the presence of sulfur (in H_2S form) and no appreciable change in stability over time. Thus a sulfur slip in the fuel is not expected to drastically affect the stack performance or endurance.

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